

**REMARKS****Claim Status**

Claims 1-27 are pending in the application. Claims 22-27 were withdrawn by the Examiner, as being drawn to a non-elected invention. Claims 1-3, 7-10, 13-18, 20-23 and 25 have been amended. Claim 19 has been cancelled. New Claims 28-52 have been added.

**Claims Amendment**

Claim 1-3, 7-10, 13, 15, 17-18, 20-21 and withdrawn Claims 22, 23 and 25 have been amended to comply with the U.S. Patent and Trademark Office rules and practices.

Claims 1, 22 and 25 have been amended to replace the term "treatment" in step H with the term "partially hydrolyzing the polyphosphoric acid in the membrane". Support for this amendment can be found in paragraph [0075] of the original application.

In addition, Claims 1, 22 and 25 have been amended by replacing the term "obtainable" with the term "obtained".

Claims 13-15 have been amended to more particularly define the claimed invention.

Claims 16-17, 20-21 and 23 have been amended to correct an obvious typographical error.

New Claims 28-52 have been added.

New Claims 28-42, 46-47 and 50-51 are drawn to the preferred embodiments of the present invention as recited in original Claims 1, 7-10, 13, 15, 17-18 and 20-21.

New Claim 43 is drawn to an embodiment of a proton-conducting membrane prepared by the process of Claim 1, in which the final polymerization occurs prior to casting the film. Support for this amendment can be found in paragraph [0073] of the original application.

New Claim 44 is directed at a proton-conducting membrane, wherein the total amount of monomers having a low phosphoric acid affinity is up to 40% by weight. Support for this amendment can be found in paragraph [0033] of the original application.

New Claim 45 is directed at a proton-conducting membrane, wherein the preferred aromatic carboxylic acids are those recited in paragraph [0024] of the original application, the preferred aromatic carboxylic acids are those recited in paragraph [0025] of the original application and the preferred heteroaromatic diaminocarboxylic acids are those recited in paragraph [0026] of the original application.

New Claim 48 is directed at a proton-conducting membrane, wherein the total amount of monomers having a low phosphoric acid affinity is up to 40% by weight and wherein the proton-conducting membrane prepared by the process of Claim 1, in which the final polymerization occurs prior to casting the film. Support for this amendment can be found in paragraphs [0033] and [0073] of the original application.

New Claim 49 is directed at a proton-conducting membrane, wherein the preferred aromatic carboxylic acids are those recited in paragraph [0024] of the original application, the preferred aromatic carboxylic acids are those recited in paragraph [0025] of the original application and the preferred heteroaromatic diaminocarboxylic acids are those recited in paragraph [0026] of the original application and wherein the proton-conducting membrane prepared by the process of Claim 1, in which the final polymerization occurs prior to casting the film. Support for this amendment can be found in paragraphs [0024]-[0026] and [0073] of the original application.

New Claim 52 is drawn to the subject matter of cancelled Claim 19. Support for this amendment can be found in paragraph [0102] of the original application.

New Claims 28-52 read on the elected species.

Rejection of Claims 1, 9-10, 15, 17-18 and 20-21 Under 35 U.S.C. §112, Second Paragraph

Claims 1, 9-10, 15, 17-18 and 20-21 have been rejected under 35 U.S.C. §112, second paragraph, as being indefinite. The Examiner stated that the term "obtainable" in Claim 1 and the term "preferably" in Claims 1, 9-10, 15, 17-18 and 20-21 renders Claims 1, 9-10, 15, 17-18 and 20-21 indefinite.

Claims 1, 7-10, 13, 15, 17-18 and 20-21 are amended. Specifically, the phrase "obtainable" in Claim 1 has been replaced with the phrase "obtained". The recitations of the preferred embodiments in Claims 1, 7-10, 13, 15, 17-18 and 20-21 have been deleted. New Claims 28-42 have been added to be drawn to the preferred embodiments of the present invention as recited in original Claims 1, 7-10, 13, 15, 17-18 and 20-21.

In view of the above, Claims 1, 7-10, 15, 17-18 and 20-21 meet the requirements of 35 U.S.C. §112, second paragraph. Reconsideration and withdrawal of the rejection are respectfully requested.

Rejection of Claims 1-21 Under 35 U.S.C. §102(b) or, in the Alternative Under 35 U.S.C. §103(a) Over Guth

Claims 1-21 have been rejected under 35 U.S.C. §102(b) as being anticipated by or, in the alternative, under 35 U.S.C. §103(a) as obvious over Guth *et al.*, WO2002/36249 (hereinafter, "Guth").

The Examiner alleged that Guth teaches a solid polymer membrane "doped with acid". The Examiner stated that the solid polymer membrane of Guth is in the form of a single or multi-layered plastic membrane to be useful for fuel cell application (title, abstract, and lines 1-24 of Guth). The Examiner alleged that such acid-doped membranes comprise at least one layer A of a polymer mixture consisting of "two or more" polymer components: (A) at least one polyazole having azole unit formula (1A) and/or (1B), and (B) a polysulfone comprising sulfone units of (2A)-(2G) and no sulfonic acid. Furthermore, the Examiner stated that Guth, at least in some cases, uses a mixture of two different polyazoles as starting material for (A). The Examiner alleged that Guth is silent about one feature "only": "such a mixture of two different polyazoles (made from A/B and C/D) are separately prepared and then combined". Therefore, the Examiner concluded that Guth and the present invention recite exactly the same mixture of two different polyazoles and stated that, since the acid-doped membrane of Guth is used for fuel cell application, a reasonable basis exists to believe that the final acid-doped membrane products of the invention inherently possess the same properties and functions.

Applicants respectfully disagree with the Examiner's conclusions. Applicants submit that the invention defined in Claims 1-21 and in new Claims 28-52 is novel and non-obvious over Guth for the reasons discussed in details below.

Applicants' invention

Applicants' invention is a new class of proton-conducting polymer membrane based on polyazole block copolymers, obtained in one embodiment, by the process recited in Claim 1. In alternative embodiments, the present invention is defined in Claims 43-45, 48-49 and 52. The materials defined by independent Claims 1, 43-45, 48-49 and 52 comprise block copolymers such that different blocks have different and predetermined phosphoric acid affinity. The polymer blocks are formed from monomers obtained by mixing one or more tertaamino compounds with one or more aromatic carboxylic acids. The tertaamino compounds and the

aromatic carboxylic acids are chosen based on their phosphoric acid affinity, so that the monomers, and therefore polymer blocks, possess predetermined, *high or low* phosphoric acid affinity. The block copolymer defined by Claims 1, 43-45, 48-49 and 52 is produced by forming two pre-polymers first (steps (B) and (D)), followed by co-polymerizing these pre-polymers (step (G), or step (F) in Claim 43).

The instant specification teaches which of the tetraamino compounds and aromatic carboxylic acids used in the preparation of the proton-conducting polymer possess high and which tetraamino compounds possess low phosphoric acid affinities (see page 4, lines 6-39 and page 5, lines 1-12). Furthermore, the instant specification teaches how to obtain polymer blocks of a desired phosphoric acid affinity (page 6, lines 15-20).

The proton-conducting polymer membrane of Claims 1, 43-45, 48-49 and 52 possess improved and unexpected properties due to its unique structure, having blocks of different and predetermined phosphoric acid affinities, as described in details below.

#### Teachings of Guth

Independent Claims 1, 43-45, 48-49 and 52 are product-by-process claims. According to M.P.E.P. §2113:

The structure implied by the process steps should be considered when assessing the patentability of product-by-process claims over the prior art [...] (*Emphasis added.*)

In *In re Garnero*, 412 F.2d 276, 162 USPQ 221 (CCPA 1979), which is quoted in M.P.E.P. §2113, the court gave clear guidelines for examination of a product-by-process claim:

The correct inquiry [...] is whether the product defined by claim 1 is patentably distinguishable over the [cited references] in view of the structural limitation [...] (*In re Garnero*, 412 F.2d 276 at 279.) (*Emphasis added.*)

In other words, for the product-by-process Claims 1, 43-45, 48-49 and 52 to be patentably distinct over Guth, it is only required that the structure formed as a result of performing the steps recited in those claims (*i.e.*, a proton-conducting polymer membrane) be different from and non-obvious over that of Guth.

Based on Applicants' Attorneys analysis of the English-language Abstract and the German-language specification of Guth, Applicants' Attorneys submit that the proton-conducting polymer membrane of independent Claims 1, 43-45, 48-49 and 52 is structurally different from membranes disclosed in Guth for the following reasons. Contrary to the Examiner's allegations, Guth does not teach a block copolymer as instantly claimed. There are no teachings in Guth of formation of two individual blocks of polyazole polymer that have a predetermined and different affinity towards phosphoric acid, which are then combined to form a block copolymer as claimed in independent Claims 1, 43-45, 48-49 and 52. At best, the resulting polyazole polymer of Guth is a so called "random" polymer, characterized by a random distribution of monomers along the polymer chain. Thus, in contrast to the pending Claims 1, 43-45, 48-49 and 52, Guth does not teach selection of different polymer blocks that have different and predetermined phosphoric acid affinities. In addition, Guth does not disclose that the individual blocks have a molecular weight corresponding to an intrinsic viscosity of up to 1.5 dl/g. In view of the forgoing, independent Claims 1, 43-45, 48-49 and 52 and claims dependent thereon are novel over Guth.

Furthermore, Guth does not render Claims 1, 43-45, 48-49 and 52 obvious. Nothing in Guth teaches or suggests the use of a polyazole block copolymer of different and predetermined phosphoric acid affinity.

The use of block copolymers, such that the blocks have different and predetermined phosphoric acid affinities results in unexpected advantages, such as high conductivity and improved mechanical properties (paragraph [0082] of the instant specification). These improved characteristics of the block copolymer membrane are the results of selecting and combining polymer blocks of different phosphoric acid affinity and would not be predictable based on the teachings of Guth.

To further support the assertion that the proton-conducting membrane defined by Claims 1, 43-45, 48-49 and 52 possess unexpected, advantageous properties that would not be predictable based on the teachings of Guth, Applicants submit herewith Exhibit A. Exhibit A is a copy of relevant parts from the doctoral thesis of Eugene Scanlon, Ph.D, one of the named inventors of the present application. The doctoral thesis of Eugene Scanlon is publicly available from Rensselaer Polytechnic Institute, Department of Chemistry, Libraries' Catalog:

Location: Folsom Thesis Stacks (3<sup>rd</sup> Floor) & Folsom Storage (Circulation Desk)

Call #: 2005

Author: Scanlon, Eugene

Title: Polybenzimidazole based segmented block copolymers for high temperature fuel cell applications;

Published: 2005.

The relevant parts of Dr. Scanlon's thesis (Chapter 3, submitted as part of Exhibit A) are summarized in the Advantages of the Applicants' Invention section of this document, bellow.

#### Advantages of the Applicants Invention

The experiments described in Exhibit A demonstrate that films prepared from block copolymers possess higher conductivity (due to retention of phosphoric acid) and have higher mechanical strength (as measured by stress at break and strain at break) than the films made from homopolymer blends (Exhibit A, Section 3.2.3, page 80) or random copolymers (Exhibit A, Section 3.2.4, page 81).

The polyazole block copolymers of the independent Claims 1, 43-45, 48-49 and 52 correspond in structure to the "segmented block copolymers", described in Exhibit A, Section 3.2.5, page 81<sup>1</sup>. The "segmented block copolymers" of Exhibit A and the block copolymers of the instant application were prepared according to the same procedure, *i.e.* by pre-selecting polymer blocks of different phosphoric acid affinity. As described in details on page 81, Section 3.2.5 of Exhibit A, block copolymers were prepared by mixing isophthalic acid (IA) and 3,3'-4,4'-tetraaminodiphenyl sulfone (TAS) in one resin flask, while terephthalic acid (TA) and 3,3'-4,4'-tetraaminobiphenyl (TAB) were added in equimolar amounts to a separate flask.

As described in the instant application, TAS has high phosphoric acid affinity, TAB has low phosphoric acid affinity, while both IA and TA have low phosphoric acid affinity. Therefore, combining TAS and IA to form meta-sulfone PBI (m-SPBI), produces block of high phosphoric acid affinity, while combining TAB and TA to get para PBI (p-SPBI), produces block of low phosphoric acid affinity. Combining m-SPBI and p-SPBI blocks forms polyazole block copolymer in which different blocks have different phosphoric acid affinities. Hence, the

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<sup>1</sup> The term "segmented block copolymer" used in Exhibit A is interchangeable with the term "block copolymer" of the instant application.

“segmented block copolymer” described in the Exhibit A correspond in structure to the polymer of Claims 1, 43-45 and 48-49.

“Homopolymer blends” (Exhibit A, Section 3.2.3, page 80) were prepared by mechanically mixing previously prepared para-PBI (*p*-PBI) and meta-sulfone PBI (*m*-SPBI) polymers.

“Random copolymers” of Exhibit A were prepared by adding IA and TAS in equimolar ratios and terephthalic acid (TA) and 3,3'-4,4'-tetraaminobiphenyl (TAB) also in equimolar ratios to a reaction flask (Exhibit A, Section 3.2.5, page 81). Subsequently, PPA was added to the reaction flask. At best, the resulting polyazole polymers of Guth are so called “random copolymers”, characterized by a random distribution of monomers along the polymer chain.

*Titration experiment (Exhibit A, Section 3.3.6.2, pages 107-110)*

To determine the retention of PA and therefore conductivity of films made from different copolymers, titration experiments were performed, as described below.

Titration experiments were performed using the films cast from the three samples: random copolymers, homopolymer blends and block copolymers, described above. Each polymer film was evaluated by titration of three samples with sodium hydroxide. The results of titration experiments are shown in Table 3-6 (Exhibit A, Section 3.3.6.2, page 108) for the films hydrolyzed under two sets of experimental conditions: low humidity (22-25% relative humidity) and high humidity (60% relative humidity)<sup>2</sup>. Table 3-6 presents the results of measurements of phosphoric acid content (moles of PA per mole of polymer repeat unit) and phosphoric acid concentration (weight percent) in the films made from copolymers, random copolymers or homopolymer blends. As can be seen from Table 3-6, after hydrolysis at 25% relative humidity, the films made from block copolymers contained 91.5 moles of PA per repeat unit in comparison to 10.2 moles of PA per repeat unit for random copolymer films, and 54.7 moles of PA per repeat unit for homopolymers blend. After hydrolysis at 60% relative humidity, the block copolymer films contained 34.6 moles of PA per repeat unit in comparison to 17.4 moles of PA per repeat unit for random copolymers and 26.5 moles of PA per repeat unit for homopolymers blend. The results of titration experiments are also shown graphically in Figure 3-11, page 109 of Exhibit A. As can be seen from Figure 3-11, the films prepared from the block copolymers

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<sup>2</sup> Atmospheric moisture causes hydrolysis of both phosphoric acid and the membrane.

retained more PA when compared to the films prepared from random copolymers and homopolymer blends, regardless of relative humidity. The concentration of phosphoric acid in the film is in direct correlation with the conductivity of the film, as is well known to one of ordinary skill in the polymer art. Therefore, the films prepared from block copolymers, which are the best at acid retention, also have the highest conductivity.

*Mechanical properties (Exhibit A, Section 3.3.6.3, pages 111-113)*

To determine mechanical properties of films made from block copolymers, random copolymers and homopolymer blends, "stress at break" and "strain at break"<sup>3</sup> of the membrane prepared from each type of the material were measured. Average values for "stress at break" and "strain at break" for all three samples are given in Table 3-7, page 112 of Exhibit A. The experiments were performed using three samples from the same polymerization / film lot used in the above-described titration experiments. Experiments were performed under two sets of conditions: low relative humidity (22%-25%, at 22°C) and high relative humidity (60%, at 22°C).

These experiments show that films made from the block copolymers had the best mechanical properties, regardless of the experimental conditions. Specifically, at lower relative humidity (22-25%), films made from copolymers had "stress at break" of 1.2-1.8 MPa and "strain at break" of 400-525% strain at break, in comparison to 1.0-1.4 MPa and 75-175% for random films and 0.2-0.3 MPa and 20-55% for films made from homopolymer blends. At higher relative humidity (60%), films made from block copolymers had 1.5-1.7 MPa stress at break and 700-875% strain at break. The films made from homopolymer blends were too weak to be held by the tensile testing instrument and could not be tested and films made from random copolymers had 0.7-0.8 MPa "stress at break" and 325-350% "strain at break".

In summary, regardless of experimental conditions, block copolymers showed the best "strain at break" and "stress at break" when compared to homopolymer blends and random copolymers.

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<sup>3</sup> The "tensile stress" is well known to one of ordinary skill in the polymer art as stress applied to extend the test piece, calculated as a force per unit area of the original cross-section of the test length, while "stress at break" is defined as the "tensile stress" at which the test piece ruptures. Units of "stress at break" are MPa. The "tensile strain" is also well known to one of ordinary skill in the polymer art as the relative elongation of a material when subjected to tension, while the "strain at break" is defined as "tensile strain" at which the test piece ruptures.



In the view of the above data, proton-conducting membranes that result from performing the steps recited in Claims 1, 43-45, 48-49 and 52 are different from and unexpectedly better than the materials disclosed in Guth. As such, Claims 1-21, as well as, new Claims 28-52 are novel and non-obvious over Guth. Reconsideration and withdrawal of the rejection under 35 U.S.C. §102(b) or, in the alternative, under 35 U.S.C. §103(a) over Guth are respectfully requested.

Rejection of Claims 1-21 Under 35 U.S.C. §103(a)

Claims 1-21 have been rejected under 35 U.S.C. §103(a) as being unpatentable over Sakaguchi *et al.*, U.S. 7, 288, 603 B2 (hereinafter, "Sakaguchi") in view of Narang *et al.*, WO 01/94450 A2 (hereinafter, Narang) or Guth.

The Examiner stated that Sakaguchi teaches that in the course of making acid-doped proton-conducting polymer membrane, polyazole type polymer can be readily prepared by directly heating the precursor mixtures including (A) amine compound and (B) carboxylic acid compound. The Examiner acknowledged that Sakaguchi does not teach using a mixture of two different polyazoles separately prepared, and then combined. However, the Examiner alleged that two other references, Narang and Guth in combination or alone can teach such a subject matter.

The Examiner stated that Narang teaches using a binary polymer blend optionally including a third polymer in making acid-doped or acid-containing polymer membranes for using in fuel cell. With regard to Guth, the Examiner repeated his arguments that Guth teaches some solid polymer membranes "doped with acid" and that at least in some cases, Guth uses a mixture of two different polyazoles as starting material for (A).

The Examiner concluded that a person of ordinary skill in the art would have found it obvious to modify Sakaguchi's process of making an acid-doped proton-conducting membrane by using the mixture of two different polyazoles that are taught by Narang and Guth.

Applicants respectfully disagree with the Examiner's statements and conclusions.

As acknowledged by the Examiner, Sakaguchi does not disclose a mixture of two different polyazoles, having different affinities towards phosphoric acid. In addition, Sakaguchi neither teaches nor suggests that using such mixture would result in the proton-conduction membrane having beneficial properties. Sakaguchi does not teach or disclose any problem with

the process of preparing the proton-conducting membrane that would benefit from such modification. Based on the teachings of Sakaguchi, one of ordinary skill in the art would not be motivated to use the mixture of two different polyazoles, instead of single polyazole.

Even if one of skill in the art would modify Sakaguchi's process by using the mixture of two different polyazoles that are taught by Narang and Guth, one would still not arrive at the instant invention, as claimed in independent Claims 1, 43-45, 48-49 and 52. Neither Narang nor Guth teaches a block copolymer having blocks of different affinities, as discussed above with regard to the rejection of Claims 1-21 over Guth. There are no teachings in either Guth or Narang of formation of two individual blocks of polyazole polymer that have a predetermined and different affinity for phosphoric acid, and then combining these blocks to form a block copolymer, as claimed in independent Claims 1, 43-45, 48-49 and 52. At best, the resulting polyazole polymers of Guth and Narang are so called "random" polymers, characterized by a random distribution of monomers along the polymer chain.

Furthermore, neither Guth nor Narang discloses that the individual blocks have a molecular weight corresponding to an intrinsic viscosity of up to 1.5 dl/g. Both Guth and Narang are silent on any difference in the affinities of different blocks of polymer towards the phosphoric acid.

Additionally, block copolymer membranes defined in Claims 1, 43-45, 48-49 and 52 have improved mechanical properties and high conductivity. These improved characteristics of the block copolymer membrane are the results of selecting and combining polymer blocks of different phosphoric acid affinity. These superior properties of the proton conducting membranes of independent Claims 1, 43-45, 48-49 and 52 are discussed in details above.

In view of all of the above, proton-conducting membranes that are obtained by performing the steps recited in Claims 1, 43-45, 48-49 and 52 are different from and unexpectedly better than the materials disclosed in either of the above-cited references alone or in combination. As such, Claims 1-21, as well as, new Claims 28-52 are novel and non-obvious over Sakaguchi in view of Narang or Guth. Reconsideration and withdrawal of the rejection are respectfully requested.

**Information Disclosure Statement**

A Supplemental Information Disclosure Statement (SIDS) is being filed concurrently herewith. Entry of the SIDS is respectfully requested.

**CONCLUSIONS**

In the view of the above, the proton-conduction membrane of Claim 1 possess unique and advantageous properties that could not have been predicted based on any of the cited references individually or in combination, hence the proton-conducting membrane produced by steps recited in Claim 1 is a special technical feature and Groups I-III relate to a single general inventive concept under PCT Rule 13.1. Applicants respectfully request reconsideration and withdrawal of restriction requirement.

Respectfully submitted,

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